

Poisoning of Heat Pipes

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Abstract. Thermal management is critical to space exploration efforts. In particular, efficient transfer and control of heat flow is essential when operating high energy sources such as nuclear reactors. Thermal energy must be transferred to various energy conversion devices, and to radiators for safe and efficient rejection of excess thermal energy. Applications for space power demand exceptionally long periods of time with equipment that is accessible for limited maintenance only. Equally critical is the hostile and alien environment which includes high radiation from the reactor and from space (galactic) radiation. In space or lunar applications high vacuum is an issue, while in Martian operations the systems will encounter a CO₂ atmosphere. The effect of contact at high temperature with local soil (regolith) in surface operations on the moon or other terrestrial bodies (Mars, asteroids) must be considered.

Introduction

Contamination, or poisoning, of the working fluid and variations in residual gas pressure can significantly alter the working fluid evaporation and heat transport rates. Possible sources of such contamination include dissolution of containment and wicking material by the working fluid, diffusion of low level impurities dissolved in the materials, penetration and solution of gases, and reaction with materials such as the regolith minerals from the surrounding environment. Such poisoning can change the surface tension properties of the working fluid. Long term extensive testing in well simulated space situations is needed prior to committing a final design for heat pipes for missions of several years in duration. In this work the intent is two-fold, namely (1) to initiate experiments to evaluate the diffusion of candidate gases through Inconel 625, a high temperature candidate containment material, and (2) to evaluate reactivity with a lunar regolith simulant. The composition of Inconel 625 is shown in Table 1.

Description of Research

The thrusts of the preliminary part of the research have been, first to evaluate and measure the permeability, solubility and diffusion of gases through Inconel 625. The second series of tests is designed to discern if there is any reaction of Inconel 625 with regolith simulant materials.

Diffusion of gases through Inconel 625

Penetration of gases through metals can be a problem over a long time period. The penetration of hydrogen and helium through Inconel 625 samples was examined using a system modified from equipment originally designed to measure diffusion out of fused silica for semiconductor crystal growth.¹ The measuring system is shown schematically in Figure 1. A segment of Inconel 625 tube (30 cm in length, 16 mm OD, and 260 μ m in wall thickness), sealed at one end, is connected to a barocel gauge and, through a valve, to a vacuum system. The test tube was enclosed by an external tube (Inconel 625 with 508 μ m wall thickness), connected at one end to hydrogen and argon tanks (9N purity each), and to a bubbler or vacuum at the other

end. Measurements of both permeability and solubility are made after initial saturation of the material with diffusant to achieve steady state was reached.

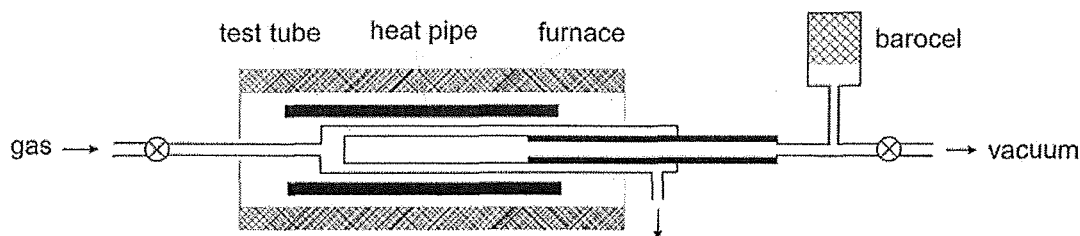


FIGURE 1. Permeation/Solubility Measurement System

TABLE 1. Composition Range for Inconel 625

	Minimum	Maximum
Carbon		0.10
Nickel	balance	balance
Chromium	20.0	23.0
Iron		5.0
Silicon		0.5
Manganese		0.5
Sulfur		0.015
Molybdenum	8.0	10.0
Titanium		0.4
Cobalt		1.0
Niobium + Tantalum	3.15	4.15
Aluminum		0.4

The results of solubility at 1 atm., obtained from the amount of hydrogen desorbed from the sample, are shown in Figure 2. For comparison, solubility of hydrogen in nickel (Fromm and Hörz, 1980)² is also shown. As follows from Figure 2, solubility of hydrogen in Inconel 625 is about an order of magnitude greater than that in pure nickel. The results of the permeability measurements are shown in Figure 3. Each experimental point was calculated from an average of several measurements of the rate of pressure increase in the system. The inset shows the raw data which demonstrates that the permeation rate at 0.089 atm of H₂ is about one order of magnitude lower than that at 1 atm. This suggests at least a partially molecular mechanism of solution of hydrogen in Inconel 625 in our temperature range. Further interpretation of the permeability results, in particular on the effect of H₂ pressure, requires additional studies. Based on SIMS analysis of selected samples and numerical simulations of diffusion, the permeability of thermal He in Inconel 625 at 1040°C has been estimated to be lower than 10⁻¹⁸ cm²/s. These results indicate a very low penetration rate, and lead to a conclusion that there would be little or no penetration of helium through Inconel 625 of any practical thickness at the operating temperature (typically ~700 °C) over a period of several years.

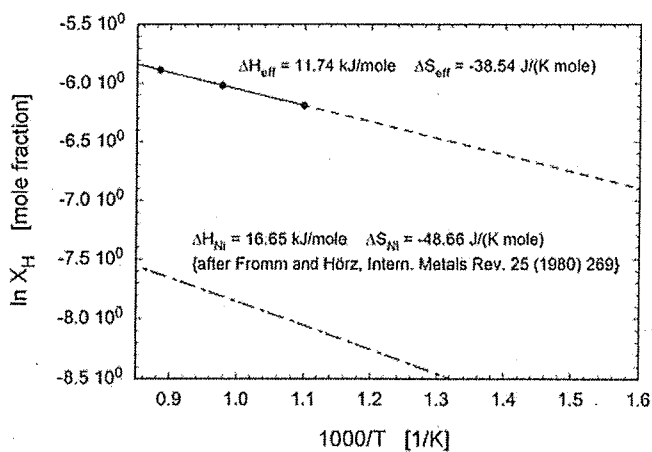


FIGURE 2. Solubility of Hydrogen in Inconel 625 at 1 atm

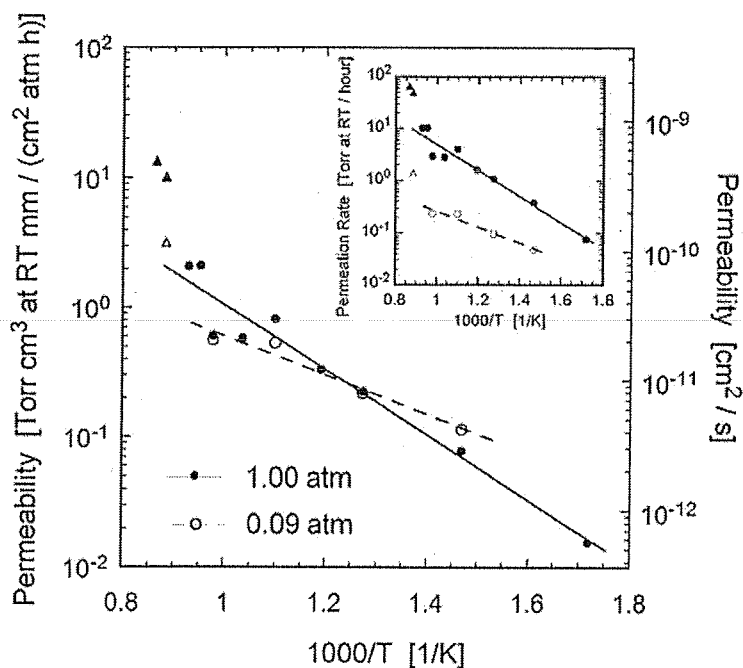


FIGURE 3. Permeability of hydrogen through Inconel 625. Inset is the permeation rate.

Reactivity of JSC-1 Lunar simulant with Inconel 625

In the absence of lunar soil, the well characterized JSC-1 simulant material was used in tests to determine if and how it would react with Inconel 625 at elevated temperatures. The composition of JSC-1 is given in Table 2 and is compared to a lunar mare soil from Apollo 14. The simulant is prepared by ball milling to 200 mesh, sieving, and then heating in an oven at 150°C for a week to drive off moisture. Initially the plan was to place coupons of Inconel 625 in fused silica ampoules with the simulant, evacuate them and then heat them over periods of time to determine reactivity. However, the simulant devitrified the fused silica at 875°C, and the use of silica as a containment vessel was abandoned. The technique has been modified so

TABLE 2. Major Element Compositions for JSC-1

	JSC-1 (Mean of 3)	Lunar Soil 14163
Oxide	Wt.%	Wt.%
SiO₂	47.71	47.3
TiO₂	1.59	1.6
Al₂O₃	15.02	17.8
Fe₂O₃	3.44	0
FeO	7.35	10.5
CaO	9.01	9.6
MgO	10.42	11.4
Na₂O	2.7	0.7
K₂O	0.82	0.6
MnO	0.18	0.1
Cr₂O₃	0.04	0.2
P₂O₅	0.66	
LOI	0.71	
Total	99.65	99.8
	LOI – Loss on Ignition	

that the simulant is loaded directly into ¼" diameter Inconel tubes in a glove box. Prior to fabrication the Inconel stock is cleaned internally and externally and then inspected by a specific NASA cleanliness specification. The glove box operates with both water vapor and residual oxygen at levels of 1 ppm or lower. The procedure is demonstrated in Figures 4b and 4b.

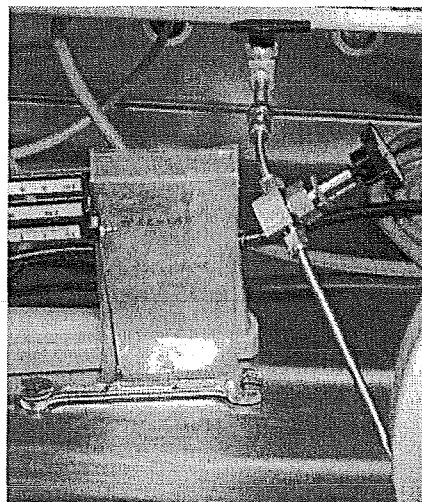
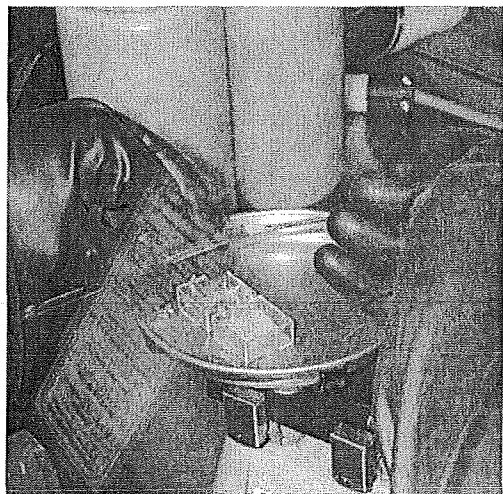


FIGURE 4. (a-left) simulant material being weighed under 1 atm argon in the glovebox, and (b-right) the Inconel tube attached to the vacuum system prior to removal for TIG welding

After loading under approximately 1 atmosphere of argon, the tube is attached to the vacuum line shown in figure 4b and the valve opened to pump out the argon. The samples are then crimped on the inside of the valve, sealed in plastic bags and then transferred for TIG welding.

Thermal treatments consist of heating in a horizontal tube furnace for periods of one to three weeks and at temperatures of 750°C, 875°C, 950°C, and 1000°C. As the furnaces are run in air, there is oxidation on the surface of the Inconel tubes. Isothermal furnace liners made from the same alloy have been run successfully in our laboratory in air at temperatures up to 1100°C without problems. A post-treatment photograph of the tube is shown in Figure 5; the oxidation does not affect the integrity of the tube. The TIG welds can be seen. The tube is shown after being opened, and the heat treated simulant has been removed. While not obvious in this photograph, there is some "clinkering" of the material. The sintered lumps are presumably formed from liquid phases produced during the heat treatment. As shown in Table 1 the simulant contains over 3% alkali oxides which would result in liquid forming at temperatures below 900°C. The powder is then examined by x-ray diffraction in a Rigaku D-MAX powder diffractometer using Cu K radiation and Materials Data Incorporated software for data collection and interpretation.

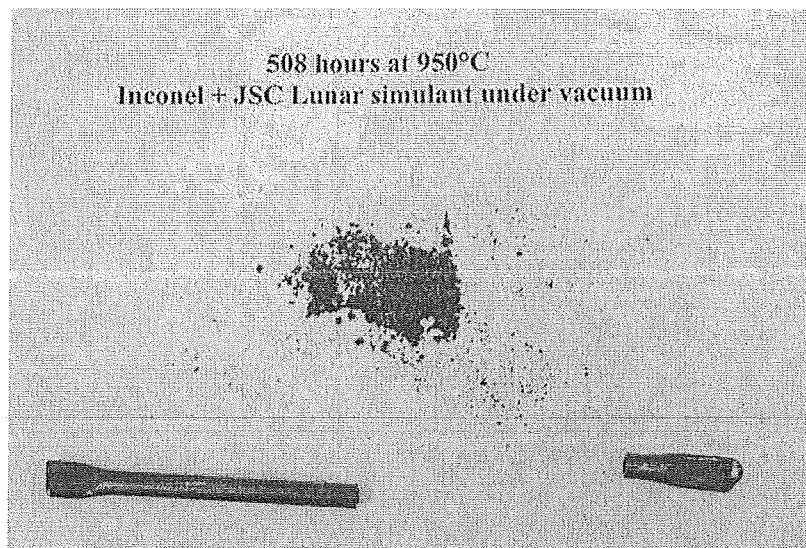


FIGURE 5. Post-heat treatment simulant, after opening the Inconel tube

Results – X-Ray Diffraction

A powder diffraction pattern of JSC-1 is shown in figure 6. The pattern shows a complex series of diffraction lines, as normally found with mixtures of minerals. The glass phase has contributed to the background hump at 25-30° 2θ, and at angles below 10° 2θ (not shown). Comparison of the strongest lines with known data bases enables the pattern to be identified as mainly anorthite ($\text{CaAl}_2\text{Si}_2\text{O}_8$), with some albite ($\text{NaAlSi}_3\text{O}_8$). This is a typical plagioclase feldspar and confirms EPMA work done at Marshall Space Flight Center.⁴ Following treatment at 950°C, the basic pattern remains similar, but there are subtle differences. The post 950°C treatment sample pattern is shown in Figure 7. Of specific interest is the peak at 18.3° 2θ. This corresponds to a major peak in chromite, a spinel containing iron, chromium and oxygen, but no silicon. Chromite is a high symmetry mineral with very few peaks. All others would be masked by the plagioclase pattern. A very small amount of chromite is present in JSC-1, and at 18.3° 2θ the 111 reflection may be present in the untreated material as in Figure 6. The stimulant is believed to contain approximately 98 ppm of Cr. This evidence is not sufficient to confirm that there is additional chromite in heat treated samples, and it is intended to pursue other techniques be used to investigate this possibility.

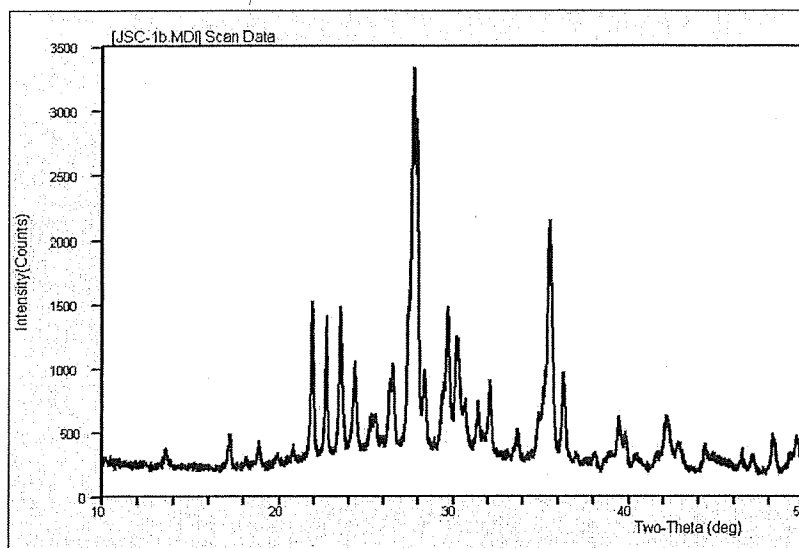


FIGURE 6. JSC-1 simulat. X-Ray powder diffraction pattern taken with Cu K radiation

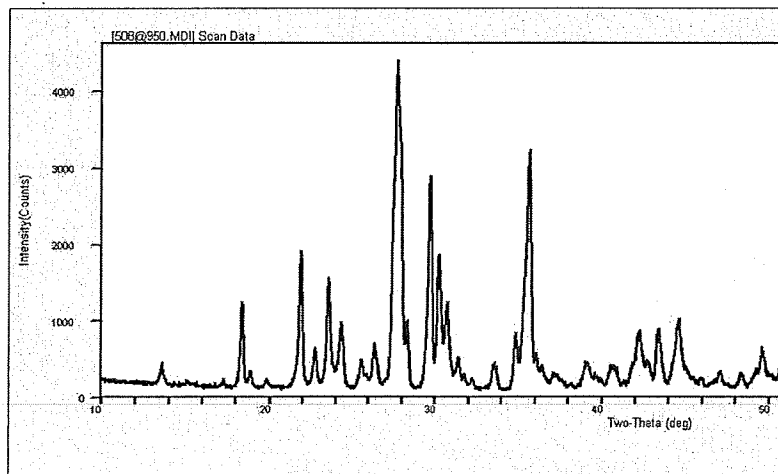


FIGURE 7. X-Ray diffraction pattern from JSC-1 heated for 508 hours at 950°C under vacuum in Inconel 625

Results - Electron Microprobe Analysis

In order to examine the material that is close to the walls and which may have reacted with the Inconel 625, the tube is sectioned through the center. The curvature of the tube makes focusing difficult in an optical microscope, and so the venerable procedure used to make transmission electron microscope replicas is used to image a flatter surface. This involves putting drops of acetone on the inside of the tube and then covering with a special plastic which is allowed to partially dissolve in the acetone. An intimate contact with the wall surface is formed, which faithfully replicates the surface morphology. Following the evaporation of the acetone, the plastic is removed, and with it come particulate matter from the wall surface. Very few particles were in fact obtained, insufficient for powder diffractometry; to examine these in the scanning electron microscope, the plastic is dissolved in acetone, leaving the particles in a filter paper. Double sided carbon tape is then used to gather the particles from the filter paper so that they can be examined in the electron microprobe analyzer (EPMA) using energy dispersive analysis (EDS) for elemental identification.

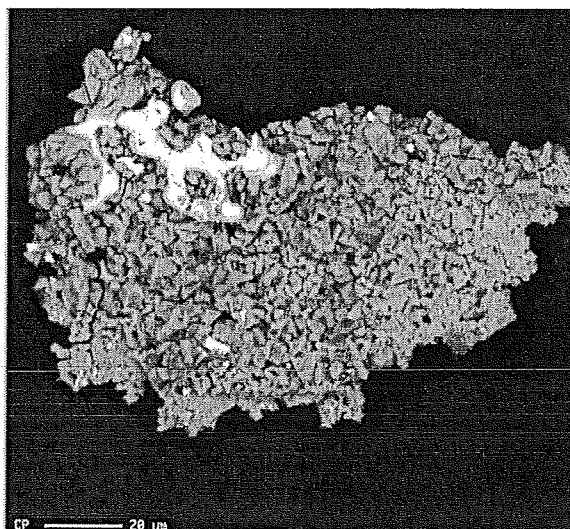


FIGURE 8. Secondary electron image of extracted particle extracted from Inconel tube

The particulate material collected from the inner surface of the Inconel tube is shown in a secondary electron photograph in figure 8. Clearly there are two major groups of phases present. This particle is one of many that showed a mixture of metal-rich (light phase), and mineral phase. Note the crystalline nature of the material, and the octahedral shape of some of the particles. Chromite is a spinel, a diamond face centered material and is known to form octahedra. Figure 9 shows the EDS pattern from the metallic phase. The spectrum shows all the Inconel elements as shown in Table 1, and also includes evidence of elements from silicate minerals (Mg, Al, Si, O). These latter are probably the result of the spread of the electron beam due to the rough surface of the sample. In comparison, the EDS pattern for the particulate material in Figure 10 shows evidence of high amounts of chromium and oxygen in addition to the expected silicon. The iron content is low, but the high chromium content indicates that the material has locally picked up chromium, presumably from the Inconel, and it adds to the possibility that the diffraction line in the bulk of the powder contains chromite.

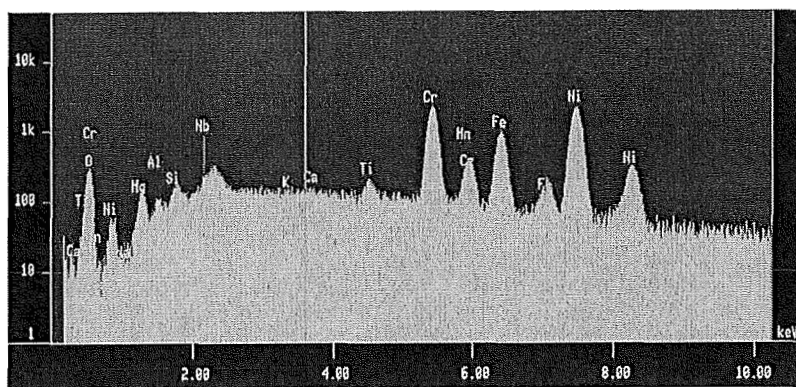


FIGURE 9. EDS spectrum of metallic (lighter) phase from figure 8.

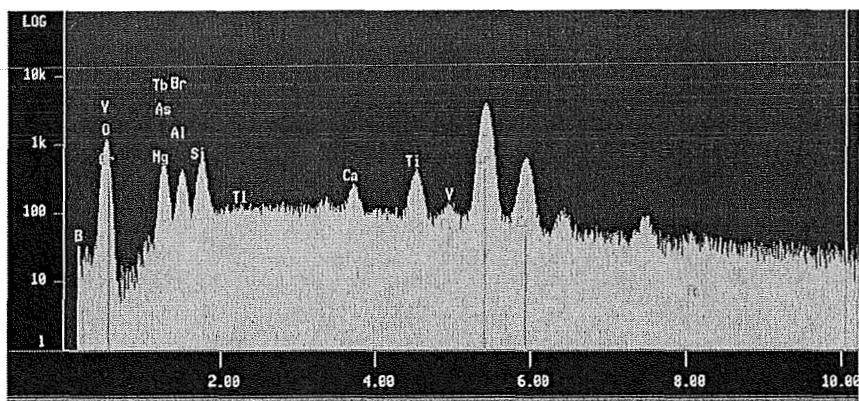


FIGURE 10. EDS pattern from the particulate matter shown in Figure 8

Discussion

There is evidence that the simulant has acquired chromium resulting in additional chromium-rich mineral phases. This can be explained with respect to the vapor pressures of the elements. Vapor pressures of relevant major elements in the Inconel 625 alloy are shown in Table 3. The data are calculated from the coefficients given in Table 2 taken from Bunshah.⁵ It can be seen that chromium has a higher vapor pressure than of any of the other major constituents, and is over an order of magnitude than nickel, and four or more times higher than that of iron at 875°C. Thus, under the vacuum conditions of this experiment, evaporation of chromium is not unexpected, and would be greater than the other major constituents of the Inconel phase. Reaction with the simulant to produce more chromium-rich minerals is more of a surprise, and could lead to possible corrosion on the lunar surface. Under typical operating conditions within the heat pipe interior, there will be high pressure (up to 1 atmosphere in the case of sodium) created by the working fluid, and the equilibrium partial pressure of chromium will be much lower. Also of interest is the fact that other possible heat pipe candidate materials such as stainless steel contain high amounts of chromium and could also be susceptible to this same problem. It should also be noted that refractory metals have much lower vapor pressures and hence would not suffer from this problem, while the application of a suitable coating might alleviate evaporation problems in the chromium containing alloys.

TABLE 3. Vapor Pressures of elements in Inconel 625 in torr

Temperature, °C	Nickel	Chromium	Iron
750	$6.7 * 10^{-12}$	$1.9 * 10^{-9}$	$3.6 * 10^{-11}$
875	$5.0 * 10^{-10}$	$2.9 * 10^{-8}$	$3.0 * 10^{-9}$
950	$1.9 * 10^{-8}$	$3.7 * 10^{-7}$	$9.1 * 10^{-8}$

Possible Future Research

Several important questions remain unanswered. In particular the possible interaction of the sodium with the containment metal has not been studied. This can be done simply loading sodium under vacuum into the Inconel and heat treating in the same manner as has been done with the simulant. The sodium would then be extracted and analyzed, while the inner surface of the tube would be examined for any reaction. Secondly, the assembly of thermosyphons should be continued. Preliminary loading of sodium into tubes, but not thermosyphons has been done at the time of writing. Long term tests with continuous measurement of temperatures during operation of the thermosyphons would show the quantitative effects of poisoning.

Quantitative measurements and kinetic rates for the evaporation of metals from Inconel, stainless steel and refractory metal coupons are important for determining the suitability of these materials in surface power thermal control systems. Other simple tests would be done, including further X-ray diffraction and EPMA on the treated powders including a Mars stimulant which has been heat treated. Finally, the internal surfaces of the glass tubing from the diffusion experiments would reveal the extent of evaporation of metals from the Inconel.

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